Calibration and analysis of a Direct Contact Membrane Distillation (DCMD) model using the GLUE method

I. Hitsov[1,2], S. Van Hoey[1], L. Eykens[2], T. Maere[1], K. De Sitter[2], C. Dotremont[2], I. Nopens[1]

[1]BIOMATH, Department of Mathematical Modelling, Statistics and Bioinformatics, Faculty of Bioscience Engineering, Ghent University, Coupure Links 653, 9000 Ghent, Belgium

[2]VITO - Flemish Institute of Technological Research, Boeretang 200, 2400 Mol, Belgium

Abstract: Membrane distillation is an interesting technology to separate non-volatile inclusions from an aqueous feed stream. However, to realise market breakthrough the economics of the technology need further improvement through module design and operational optimisation. Mathematical models have proven useful to pursue this objective. In this contribution, the Generalized Likelihood Uncertainty Estimation (GLUE) method was applied to calibrate a model for direct contact membrane distillation (DCMD). The model uses the Dusty Gas Model (DGM) for the mass transfer inside the membrane and a Nusselt equation based heat transfer submodel for the channels. The analysis revealed important interactions between parameters of the membrane and also showed that a satisfactory fit could only be obtained using an adaptation of the experimentally pre-calibrated Nusselt equation.

Keywords: Membrane Distillation; simulation; Dusty Gas Model; mass transfer; heat transfer

Introduction

Membrane distillation (MD) is an interesting technology mainly aimed at separation of non-volatile inclusions from an aqueous feed stream. The principle is based on a temperature difference across a hydrophobic membrane between the feed side (T_{mf}) and the permeate side (T_{mp}) which results in different partial pressures of water vapour (P_{mf}) and (P_{mp}) (**Figure *1***). The latter is the driving force for the flux of the evaporated phase across the membrane. The use of thin membranes creates high gradients of the water vapour pressure, allowing MD to be operated at low feed temperatures, potentially allowing reuse of waste heat at moderate temperatures from other processes. However, the economics of the technology need further improvement to make it a viable technology. A MD system allows for different operational modes such as feed temperatures, flow rates, concentrations, module configurations and membranes to be incorporated. One could try to improve the performance of the system by varying the operation conditions in different modules, but this is an expensive and time consuming job. Moreover, inadequate understanding of the transport mechanisms and phenomena occurring inside the system likely results in overlooking important parameter interactions.

In this respect, mathematical modelling allows building further insight (system analysis) and optimisation of the system. Efforts have been made in the literature to model heat (semi-empirical Nusselt equations) and mass transfer (Dusty Gas Model (DGM)) (Khayet, 2011). However, current MD modelling practice often uses tortuosity as a single calibration parameter. This potentially results in a model with predictive power in only a very small region of temperatures and/or flow rates. Hence, the MD model calibration needs further investigation in order to develop a model that can be used for process optimisation. In this contribution, the GLUE method is applied to meet this objective.

Material and Methods

The experimental data used for calibration were obtained in a flat sheet lab MD setup (6x20cm). A Series of 22 experiments was performed with a commercially available Accurel PP 2E HF, 170μm membrane, pore radius 0.27μm (Membrana, GER). The mean feed and permeate temperature were varied from 40 to 60˚C and 25 to 55˚C, respectively. The flow rates of the feed and permeate were varied from 16 to 90 $l.h^{-1}$ and the resulting flux varied from of 2.5 to 17.1 $kg.m^{-2}.h^{-1}$.

A weighted sum of squared errors (wSSE) was used as a goodness-of-fit measure between the experimental and simulated fluxes. The wSSE was calculated for the total range of flow rates and temperatures (i.e. across several experiments at different conditions). The simulation was set up in such way that at least 125,000 simulations are performed and the process is stopped in case the value of the minimum wSSE is stable in the last 10,000 simulations.

The parameters chosen for analysis are summarized in **Table *1***, and each was sampled from a uniform distribution. The Nusselt equation was pre-calibrated experimentally using aluminium foil as described by Phattaranawik et al. (2003) and a relation in the form of eq. 1 was retrieved as best-performing.

$$Nu = a.Re^{b}Pr^{c} \tag{1}$$

The parameters a, b and c were chosen for calibration, however parameters a and c were calibrated separately for the feed and permeate channels. Other parameters chosen for calibration were the membrane structural parameters such as porosity (ε), tortuosity (τ) and pore radius (Rp), as well as the thermal conductivity of the membrane.

The GLUE method (Beven and Binley, 1992) is a Monte Carlo based filtering technique that samples from a previously estimated space of parameters and classifies the resulting model predictions as either “behavioural” or “non-behavioural” based on a pre-defined criterion. By recognizing the uncertainty inherent to modelling and the inability to perfectly describe all natural phenomena, the GLUE method assumes that different parameter sets can have similar behaviour in terms of model performance. In this way more than one parameter set could result in satisfactory (behavioural) model predictions and important parameter interactions can be revealed. However, the choice of cut-off value that distinguishes between behavioural and non-behavioural simulations is always subjective. In this work, simulations which result in wSSE values smaller than the minimal obtained wSSE (best fit) plus 0.7 were considered as behavioural.

Results

Based on the GLUE method, an excellent calibration of the model was found for the entire set of experiments (***Figure 2***). The maximum error obtained between the experimental and predicted flux was less than 15%. The analysis revealed that one of the most sensitive parameters of the model is the Reynolds exponent in eq. 1 as shown in ***Figure 3***. The parameter had to be reduced to 75% of its initial value in order to obtain behavioural solutions (dots below the horizontal red line). It was not expected for this parameter to have such different value from the experimental pre-calibration. Possible reasons are (1) an error in the structure of the model, residing in the fact that that the Nusselt equations are developed for a solid wall structure and the flux currents are not taken into account in their development and/or (2) an error in the experimental calibration with aluminium foil. Other parameters which proved to be sensitive were the membrane porosity and tortuosity (***Figure 4***). For this membrane it was impossible to achieve a behavioural fit with a tortuosity higher than 1.4 and porosity lower than 0.7. One could question whether values of tortuosity and porosity of 1 are unrealistic, but nevertheless result in behavioural solutions. Explanation resides in the obvious correlation between the two parameters. Clearly, an increase in tortuosity can be compensated by an increase in porosity in the model.

A representation of the parameter sensitivity (***Figure 5***) is displayed where sensitive parameters have a large area between the line of behavioural solutions (grey) and non-behavioural solutions (black), whereas for non-sensitive parameters (e.g. Pr exponent) the two lines overlap. No significant sensitivity was found for the rest of the parameters, including the membrane pore radius. This is a quite surprising finding at first sight. However, this could again be explained by now a 3 dimensional parameter interaction between the pore radius, the porosity and the tortuosity (***Figure 6***). As can be seen, the behavioural parameter combinations form a tilted plane which means that changes in one parameter are counteracted by changes in the others. Modellers should be cautious when calibrating models with highly correlated parameters.

Conclusion

The GLUE method was successfully applied to a MD model, achieving excellent calibration. The model was tuned to predict the flux in the whole range of flow rates and temperatures. The preliminary result of the study shows interesting behaviour of the Nusselt equations. Although they are frequently used in MD modelling, a possibility exists that the flux through the membrane alters the pre-calibrated coefficient values. The study also revealed that the membrane structural parameters interact in a 3 dimensional way and equally good model predictions could be achieved with different sets of membrane parameters. A thorough system analysis is key in order to properly calibrate the model and achieve high predictive power.

References

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Figures and Tables

Table 1 Parameters chosen for analysis in this study.

Parameter	Calibrated value	Range in uncertainty	Parameter	Calibrated value	Range in uncertainty
Re exp. in Nusselt number	0.72	35%	Membrane porosity	0.8	[0.5-1.0]
Pr exp. in Nu number (permeate side)	0.33	10%	Membrane tortuosity	-	[1.0-2.0]
Pr exp. in Nu number (feed side)	0.33	10%	Membrane pore radius, μm	0.27	20%
Coefficient a in Nu, (feed side)	2.5	[2.0-3.0]	Thermal conductivity (membrane matrix)	Predicted from tortuosity and porosity	50%
Coefficient a in Nu, (Permeate side)	2.5	[2.0-3.0]			

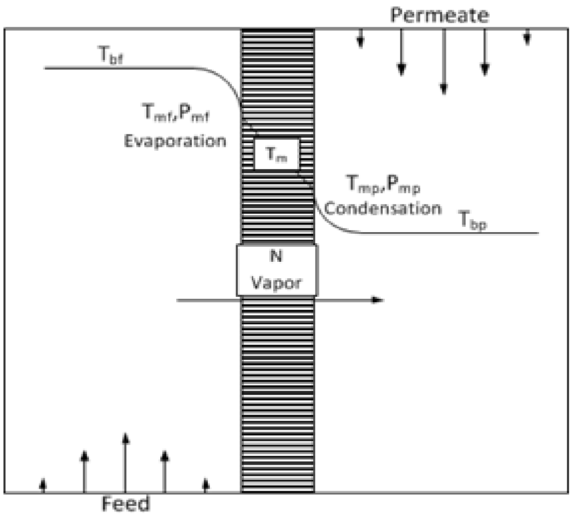

Figure 1 Operation of Direct Contact Membrane Distillation

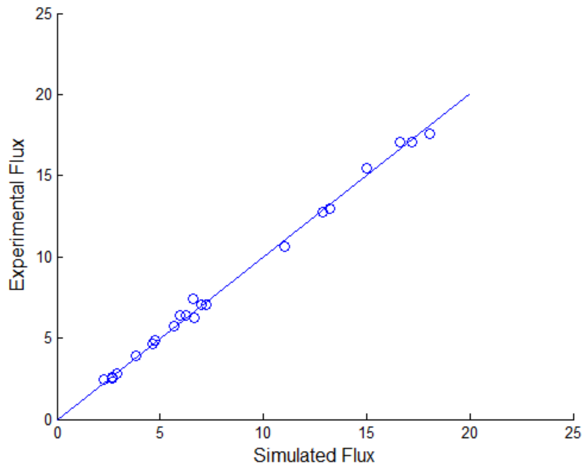

Figure 2 Outcome of the calibration performed on the model. The graph signifies the predicted versus simulated flux. XY line displayed for readability.

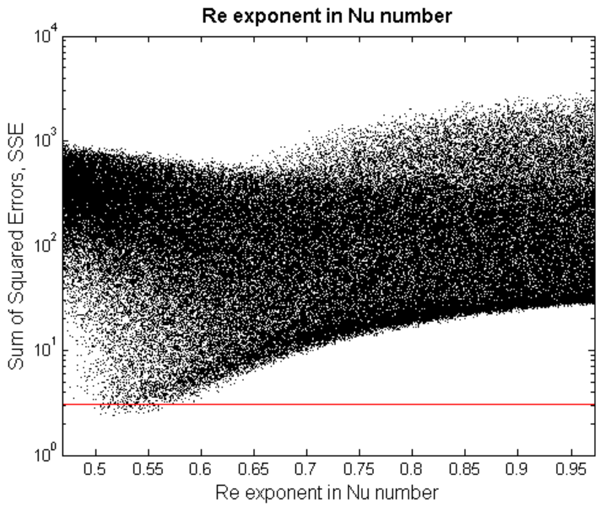

Figure 3 Scatter plot for Re Exponent in Nu number, behavioural solutions displayed as dots below the red horizontal line.

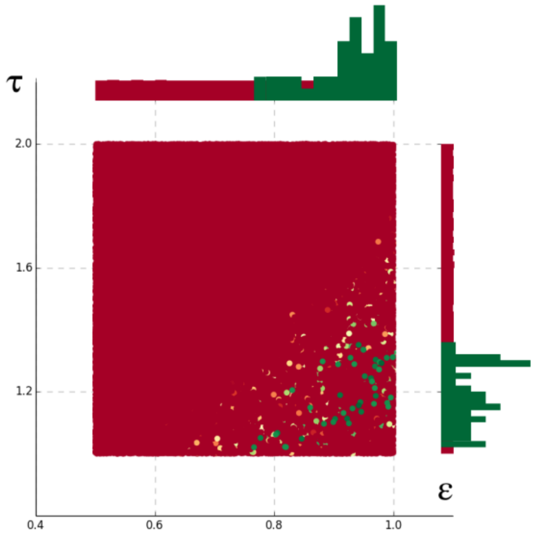

Figure 4 Scatter plot and histogram of interaction between the porosity (ε) and tortuosity (τ). Behavioural solutions displayed in green, non-behavioural in red. Gradient of colour represent solutions with fit close to the set threshold.

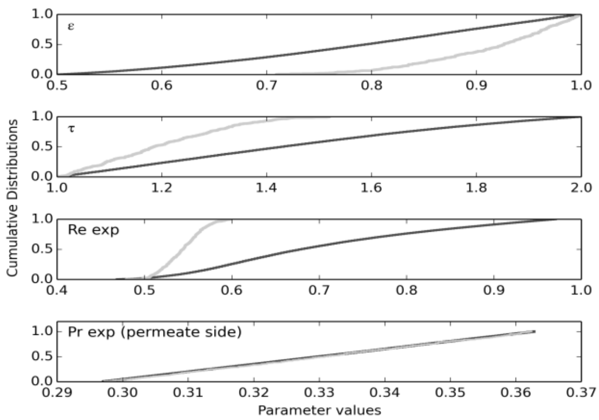

Figure 5 Cumulative distribution of behavioural (grey) and non-behavioural (black line) solutions of parameters.

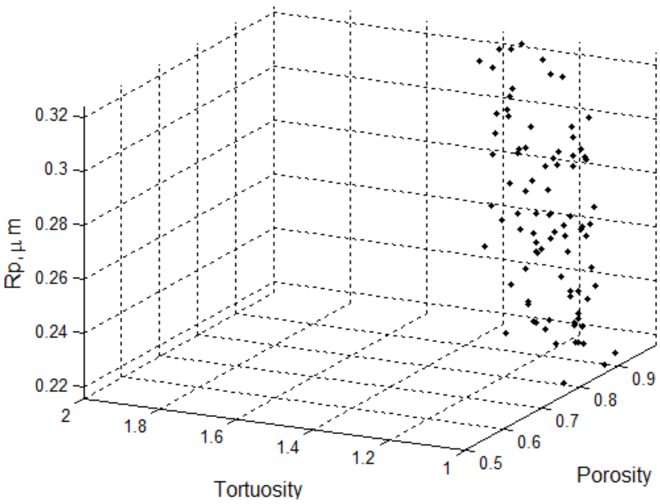

Figure 6 Three dimensional interaction between the membrane parameters. Dots represent behavioural solutions.